

Metamaterials proposed as perfect magnetoelectrics

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Magnetoelectric susceptibility of a metamaterial built from split ring resonators have been investigated both experimentally and within an equivalent circuit model. The absolute values have been shown to exceed by two orders of magnitude that of classical magnetoelectric materials. The metamaterial investigated reaches the theoretically predicted value of the magnetoelectric susceptibility which is equal to the geometric average of the electric and magnetic susceptibilities.

Magnetoelectric effect manifests a connection between electricity and magnetism and has been predicted already 1860 by Pierre Curie, but could be confirmed experimentally only about half a century ago [1, 2]. Up to now the experimentally observed effects remain rather weak. The weakness of the magnetoelectric susceptibility can to some extent be explained by the absence of strong mechanisms which couple magnetism and electricity on the microscopic level. As has been shown theoretically [1, 3], the allowed limiting value of the magnetoelectric susceptibility (ξ) is quite large and equals the geometric average of electric (χ_e) and magnetic (χ_m) susceptibilities:

$$\xi^2 \leq \chi_e \chi_m \quad (1)$$

In classical magnetoelectric materials like Cr_2O_3 the limiting value of Eq. (1) is failed by about two orders of magnitude [2, 3]. In efforts to increase the value of the magnetoelectric effect, materials revealing both strong electric and magnetic susceptibilities have been brought into consideration. Especially close to phase transitions, the electric and magnetic susceptibilities may diverge in ferroelectrics and ferromagnets. Materials simultaneously showing the ferroelectricity and ferromagnetism are called multiferroics and they are presently the subject of intensive research [2, 4, 5].

Assuming that the equality in Eq. (1) holds, it can be rewritten in the form $\xi = \sqrt{\chi_e \chi_m}$ and the constitutive relationships

$$M = \chi_m H + \chi_{me} E \quad (2)$$

$$P = \chi_e E + \chi_{em} H \quad , \quad (3)$$

can be reduced to

$$M = iP \sqrt{\frac{\chi_m}{\chi_e}} \quad , \quad (4)$$

i.e. electric polarization and magnetic moments must be directly proportional to each other. Here we have rewritten $\chi_{em} = i\xi$, and utilized the symmetry of the magnetoelectric coefficients $\chi_{me} = -\chi_{em}$ [1, 6]. The condition in Eq. (4) has been assumed in the first molecular theories for magnetoelectric effect [1] nearly a century ago.

We note that within the arguments of thermodynamic stability [6] another magnetoelectric inequality can be derived: $\xi^2 \leq \varepsilon \mu$ which utilizes the magnetic permeability $\mu = 1 + \chi_m$ and electric permittivity $\varepsilon = 1 + \chi_e$ instead of susceptibilities in Eq. (1).

Due to their unique electrodynamic properties, metamaterials (i.e. artificial materials) [7–9] may show new ways to solve the problem of the weakness of magnetoelectric coupling. Some examples of breakthroughs in various topics of modern electromagnetism which were stimulated by metamaterials are: reversing the laws of conventional optics [10], cloaking [11, 12], or overcoming the resolution limit of optical devices [13, 14]. Mixing of electric and magnetic responses is another useful property of metamaterials which can be utilized to generate new effects. As has been shown recently, the magnetoelectric coupling in metamaterials lead to strong optical activity [15–19] which is directly connected to intrinsic chirality. In agreement with the theoretical prediction [20, 21] the chirality of metamaterials can lead to giant polarization rotation and provide another routes to obtain negative refraction [22–25] for circularly polarized waves. Recently, for metamaterials with zero permittivity and permeability an estimate for the limiting value of the magnetoelectric effect $Re(\xi) \leq Im\sqrt{\varepsilon\mu}$ has been obtained [21].

Many designs of metamaterials are based on split ring resonators [26]. These elements can be seen as the smallest possible representations of the well known LC-circuit with a single inductance loop as given by the metallic ring and a tiny capacitance produced by the gap in the ring [27]. Split ring resonators have been originally developed to achieve negative magnetic permeability which is a key property for design of metamaterials with negative refraction [7]. As has been realized recently [28], the split ring resonators strongly modify the interactions with electromagnetic radiation by introducing a so called bianisotropy term into the set of basic equations [29], which is closely connected to the magnetoelectric effect [2, 5]. This additional term in the constitutive relations cross-couples the magnetic and electric fields within a split ring resonator [30, 31]. The bianisotropy offers another degree of freedom [32] in controlling the properties

of light. Here we note that, contrary to chiral metamaterials, the bianisotropy does not automatically lead to polarization rotation for geometries parallel to the principal optical axes. In order to obtain polarization rotation, these structures must be tilt or measured within off-axis geometry. The corresponding effects [33, 34] have been termed extrinsic chirality.

In this work we show that metamaterials built from split ring resonators achieve magnetoelectric effects equal to the theoretically limiting value in Eq. (1). To prove this we investigate a metamaterial of split ring resonators within different geometries, especially including those sensitive to magnetoelectricity. This allowed to obtain electric, magnetic and magnetoelectric susceptibilities and compare them with a simple circuit model. We show that the metamaterial investigated indeed reaches the theoretical limit for magnetoelectric coupling. This value is due to direct proportionality of electric and magnetic moments in split ring resonators.

Transmittance experiments at millimeter-wave frequencies ($60 \text{ GHz} < \nu < 120 \text{ GHz}$) were carried out in a Mach-Zehnder interferometer arrangement [35, 36]. This arrangement allows to measure both the intensity and the phase shift of the radiation transmitted through the sample within controlled polarization geometries. The split ring resonator arrays used in present experiments were prepared by chemical etching of copper-laminated board. The rings are typically $0.35 \text{ mm} \times 0.35 \text{ mm}$ in size with the gap width $d = 0.17 \text{ mm}$. The lattice constant of the metamaterial is $l = 0.7 \text{ mm}$. The characteristic parameters of various sets of the split ring resonators have been varied within approximately a factor of two and showed qualitatively similar results. Woven glass with a thickness of 0.56 mm was used as a non-conductive substrate. The refractive index of the substrate has been determined in a separate experiment as $n_s = 2.07 + 0.04i$.

Split ring resonators seem to represent an ideal magnetoelectric material fulfilling the condition $\chi_{me}\chi_{em} = \chi_e\chi_m$. Indeed, from the effective RLC-circuit model and simple calculations we get

$$\chi_e = nC \cdot d^2 \cdot F(\omega) \quad \chi_m = nC \cdot S^2 \frac{\omega^2}{c^2} \cdot F(\omega) \quad (5)$$

$$\chi_{em} = -\chi_{me} = nC \cdot dS \frac{i\omega}{c} \cdot F(\omega) \quad (6)$$

Here n is the density of the rings, d and C are the effective width and capacitance of the gap, S is the area of the rings, and ω is the angular frequency. We use the Lorentz substitution $F(\omega) = \omega_0^2 / (\omega_0^2 - \omega^2 - i\omega\gamma)$ where ω_0 and γ are resonance frequency and width, respectively. The symmetry of the magnetoelectric coefficients is fulfilled automatically in this model.

In order to obtain the electrodynamic parameters of the split ring resonators we have carried out the initial experiments within the experimental geometries suggested in Ref. [29] (shown in the insets to Fig. 1).

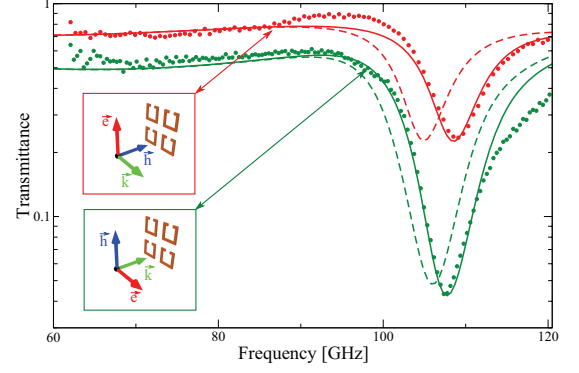


FIG. 1: Transmission characteristics of the metamaterial of split ring resonators. Two experimental geometries shown are mostly sensitive to the dielectric (green) and magnetic (red) contributions. Symbols - experiment, lines are fits using Lorentzian characteristics of the split rings. Solid lines - fits include magnetoelectric coupling, dashed lines - magnetoelectric susceptibility is set to zero, demonstrating negligible influence on the spectra. The pictograms show the geometry of the experiments.

Three relevant geometries in this case are "magnetic" (\vec{h} perpendicular to the plane of the rings), "electric" (\vec{E} parallel to the gap of the rings) and "magnetoelectric" (both excitations are realized simultaneously). In these three geometries and within reasonable approximation the effective refractive indexes basically determine the transmittance close to the resonance and they are given by: $n_e = \sqrt{\varepsilon - \xi^2/\mu}$, $n_m = \sqrt{\mu - \xi^2/\varepsilon}$, and $n_{me} = \sqrt{\varepsilon\mu - \xi^2}$, respectively. Here we neglect the influence of the substrate for simplicity. Although the magnetoelectric susceptibility is included in these equations, the dominating terms for typical parameters of the model are given by $\sqrt{\varepsilon}$, $\sqrt{\mu}$, and $\sqrt{\varepsilon\mu}$. In all cases the magnetoelectric susceptibility represents a weaker correction under the square root. This is demonstrated in Fig. 1 as we set the magnetoelectric susceptibility to zero, which simply leads to a shift of the resonance frequency. In all series of experiments with varying geometries the influence of the magnetoelectric susceptibility was below the experimental accuracy. This accuracy depends not only on experimental uncertainties, but also on the assumptions of the circuit model, like neglecting of the cross coupling effects, or assumption of infinitely small sizes of the rings compared to the wavelength. On the contrary, electric and magnetic geometries robustly depends on electric (χ_e) and magnetic (χ_m) susceptibilities. Therefore, both susceptibilities may be determined from the spectra in Fig. 1.

The result of the experiments described above may now be extended to obtain the magnetoelectric susceptibility using further geometries of the experiment. In order to get better sensitivity to the magnetoelectric susceptibil-

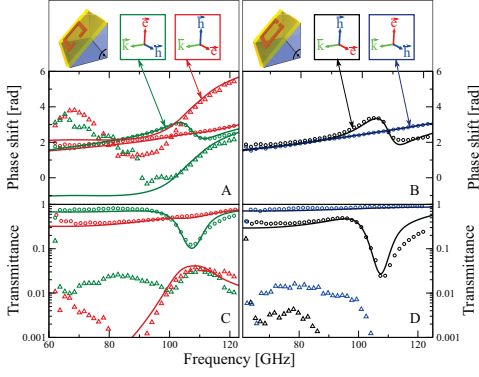


FIG. 2: Transmission and phase shift spectra of the metamaterial within various tilted geometries as indicated. Symbols - experimental data within the geometries as indicated, circles correspond to the spectra with parallel polarizers, triangles - with crossed polarizers. Solid lines are fits within the 4×4 matrix formalism [38] and using Eq. (5,6) for susceptibilities curves. In the panel C the model curves with crossed polarizers coincide for both geometries.

ity, the sample must be measured in the tilt geometry and the signals in parallel and in crossed polarizers have to be compared (see inset in Fig. 2). An example of such tilt geometries has been presented in Ref. [31], where the existence of a cross-coupling terms has been detected. The effectiveness of the tilt experiments can be demonstrated within the simplified assumptions. If we assume $ka \ll 1$, then the corresponding Maxwell equations may be solved more easily leading to analytical expressions for all relevant geometries (e.g. Ref. [37]). Here $k = 2\pi/\lambda$ is the wavevector of the electromagnetic wave and a is the sample thickness. If we further neglect the $(ka)^2$ terms compared to the terms linear in ka , then e.g. for both geometries in the left panels in Fig. 2 and within 45° incidence the following expression for the complex transmittance in crossed polarizers can be written:

$$t(\omega) = \frac{ka \cdot \xi}{2\sqrt{2}\mu + ika[1 + \xi^2 - \mu(\varepsilon + 2)]} \quad (7)$$

The last formula clearly shows that the transmittance in crossed polarizers is directly proportional to the magnetoelectric susceptibility. Therefore, this geometry provides a sensitive tool for magnetoelectric effect. In the following, the calculations have been done within the exact 4×4 matrix formalism as described in Ref. [38].

Figure 2 shows transmittance and phase shift spectra in four most important tilt geometries, both in parallel and in crossed polarizers. The main result of this data is given in the left bottom panel of Fig. 2 showing the nonzero transmittance in crossed polarizers close to the resonance of the rings (109 GHz). As expected already from the simplified equation Eq. (7), reasonably strong signal can be observed in geometries with crossed polar-

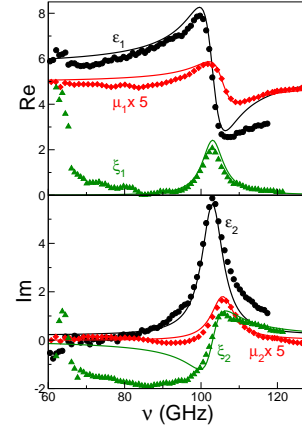


FIG. 3: Permittivity, permeability and bianisotropy of the metamaterial used in the present work. Symbols - experiment, lines - circuit model given in Eqs. (5, 6). The magnetoelectric susceptibility has been assumed to obey $\xi/\sqrt{\chi_e\chi_m} = 0.9 \pm 0.15$ instead of Eq. (1).

izers (red and green triangles). In the left bottom panel of Fig. 2 the nonzero signal below about 90 GHz for crossed geometry are due to the radiation leakage around the sample and should be neglected. The leaky signal can be also seen in the phase shifts data as shown in the left upper panel. Below about 90 GHz the amplitude of the signal is getting low and the phase measuring system loses the stability (red and green triangles).

We note another interesting feature of Fig. 2. In the geometry with ac electric fields within the plane of the rings and parallel to the gap (black circles in the right panels of Fig. 2) the resonance is clearly seen in parallel polarizers. However, we observe no signal in crossed polarizers in this geometry (black triangles). In two geometries presented in the right panels of Fig. 2 solely a stray signal could be detected. This result is supported both by simple and rigorous theories. Both calculation methods predict zero amplitude of the perpendicular polarization and reflect the symmetry of the problem.

The results of the transmittance experiments in perpendicular (Fig. 1) and tilted (Fig. 2) orientations are sufficient to determine all three complex susceptibilities for the metamaterial of split ring resonators. The results of the self-consistent calculation of these parameters based on the 4×4 matrix formalism is presented in Fig. 3. As could be already expected from the fit of the transmittance spectra, all three susceptibilities reveal a resonance-like form. The results in Fig. 3 clearly demonstrate that the metamaterial of split ring resonators indeed models a perfect magnetoelectric with $\xi^2 = \chi_e\chi_m$. Here solely a factor of 0.9 ± 0.15 had to be introduced in order to obtain the self-consistence of the data. Taking into account the assumptions made, this factor most probably reflects the uncertainties of the experiment. Finally, we recall that the condition $\xi^2 = \chi_e\chi_m$ is expected

for all materials revealing direct proportionality between electric and magnetic moments ($M = \alpha P$). Here α is a material constant which equals $\alpha = iS\omega/cd$ for split ring resonators.

In summary, using millimeter wave spectroscopy of the complex transmission coefficient the electrodynamic properties of a metamaterial of split ring resonators have been investigated. The sensitivity to the magnetoelectric effect has been obtained within tilt sample geometry and calculated within 4×4 matrix formalism. We prove experimentally and within a circuit model calculation that metamaterials from split ring resonators reach the maximum theoretical values of the magnetoelectric susceptibility limited by $\xi^2 \leq \chi_e \chi_m$. This value appears to be about two orders of magnitude above the typical coupling constants for conventional magnetoelectrics like Cr_2O_3 .

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